AN X-RAY STUDY OF COIR FIBRE

P. K. RAY AND S. B. BANDYOPADHYAY

TECHNOLOGICAL RESEARCH LABORATORIES

INDIAN CENTRAL JUTE COMMITTEE

12, REGENT PARK, CALCUTTA-40

(*Received April 20, 1965*)

ABSTRACT. The changes in spiral angle, Hermans' average angle of orientation and crystallinity with extension of coir fibre have been studied by X-ray diffraction method. It is found that the spiral angle and the angle of orientation decrease with extension and the two angles differ more in the extended state than in the unextended state and that the crystallinity increases by about 3% after extension to rupture.

INTRODUCTION

Coir is the fibre obtained from coconut husk. High extensibility (about 37%) and high lignin content (40%) of the fibre distinguish it from other cellulosic fibres. The spiral structure of the microfibrils in the secondary wall of the fibre has been studied by Astbury (1943) and Preston (1952) while Stern and Stout (1954) tried to relate the spiral angle with Herman's orientation angle. Stern (1957) has shown that high extensibility of coir fibre is chiefly because the microfibrils in the cellwall lie in perfect helical spirals extension of the fibre being related with the changes of the spiral angle, that is, the angle which a microfibril element makes with the fibre axis. He did not however examine the changes in crystallinity of the fibre although he noticed that about 4% of the extension remains as permanent even though the extended fibre is drastically treated. The object of this work is to study how far the lateral order of cellulose chains in the fibre is affected by extension. The nature of change in the spiral angle as well as the angle of orientation has also been examined.

EXPERIMENTAL

In the first series of experiments each individual fibre was extended to the maximum limit (about 30-4%) by loading it gradually on a pendulum type fibre strength tester till it broke in about a minute. The extended portions of all the fibres were then first examined for spiral angle and the angle of orientation in the form of parallel bundles and then cut to pieces for preparing randomised pellet for measurement of crystallinity. In each case unextended fibres were similarly examined under X-rays.

In another series of experiments the behaviour of the fibres at extension below maximum was checked. A bundle of coir fibre was carefully mounted on a pair of fibre clamps so that all the fibres were apparently straight. The clamps were mounted on a pendulum type cloth strength tester and load was gradually increased; the movement of the clamps was arrested after a desired amount of extension, left for 48 hours and then released, immediate relaxation being measured. It was found that the fibres retained about 80% extension. These fibres were then used for crystallinity measurement.

Spiral angle and angle of orientation : A parallel fibre bundle was kept just taut by means of a clamp in front of a collimator of 2 mm. diameter and 5 cm. length through which Ni-filtered CuK_α radiation was allowed to pass from a Philips sealed *X*-ray tube and fibre to film distance was 5 cm. An exposure of 3 hrs. with 25 KV. and 15 mA. was used. The exposure time, development technique etc. were kept as constant as possible for all the photographs.

Following Hermans (1946) *et al* and Meredith (1951) a series of microphotometer curves for (002) interferences were recorded starting from the equatorial lines of the diffraction photographs and proceeding along radial lines at angular intervals of 5°. Since the films were of low photographic density it was assumed that the X-ray intensity was proportional to the blackening and was thus linearly related to the logarithm of the intensity of the transmitted light, or of the photometer reading, since the calibration was linear over the range of measurement.

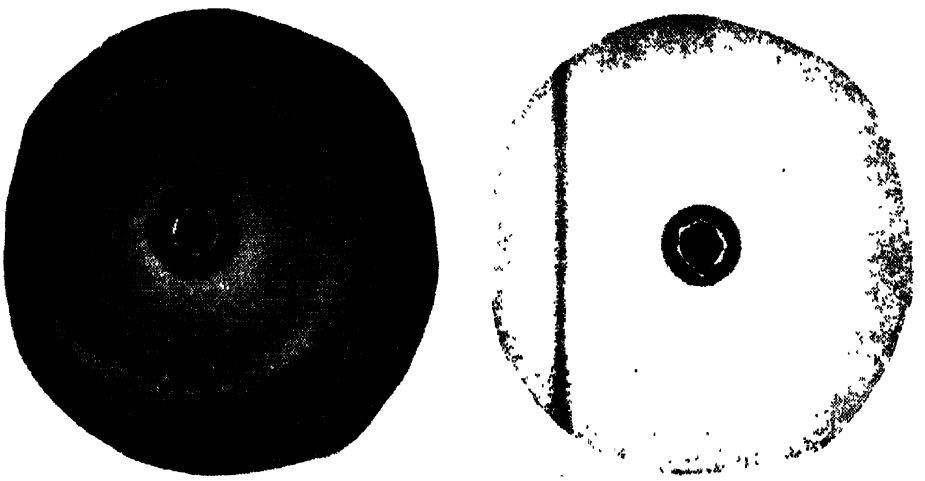
Crystallinity : As coir shows a considerably high value of extensibility it was expected that extension in coir would draw the chain molecules into parallelism and may increase the crystallinity. In finding the crystallinity the orientation effects were eliminated by a randomised preparation of the fibres forming pellets with a very small quantity of adhesive. The pellet was then introduced into a mould, a hole of ½mm. diameter drilled in a brass plate 1 cm. thick. The other end of the hole was temporarily closed by a screwed plate. A constant pressure was exerted on the pellet with a steel piston having a flat end just fitting the hole. The pellet was then taken out of the hole from the other end after unscrewing the plate. The density of the pellets was kept fairly constant by using weighed amounts of fibre in each case. The pellet was then mounted in the camera for X-ray photographs, which were taken under similar conditions.

RESULTS AND DISCUSSION

Figs. 1 and 2 are the typical X-ray photographs for parallel bundles and randomised pellets respectively, (a) and (b) indicating unextended and extended fibres.

The widely drawn out interference arcs of Fig 1 (a, b) with two maxima on either side of the equator are the special features of the photographs of fibres with spirally laid microfibrils. X-ray intensity when plotted against the angular

distance from the equator gave graphs as shown in Fig. 3(a, b). Half of the angular seperation between the maxima was taken as the vertical spiral angle.



(a) (b)

Fig. 1(a, b) X-ray photographs of parallel bundles of coir fibre: (*a*) Unextended; (*b*) Extended.

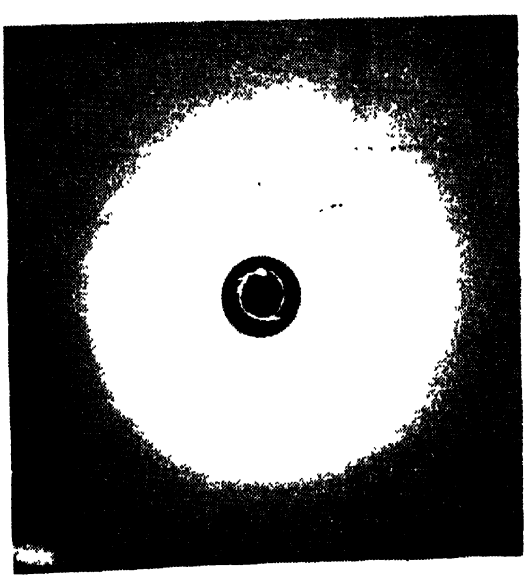


Fig. 2(a)

Fig. 2(*a*) Unextended, (*b*) Extended.

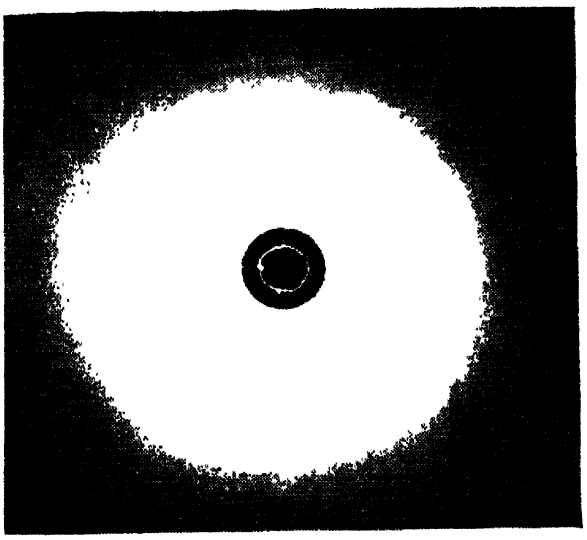


Fig. 2(b) X-ray photograph of randomised pellot :

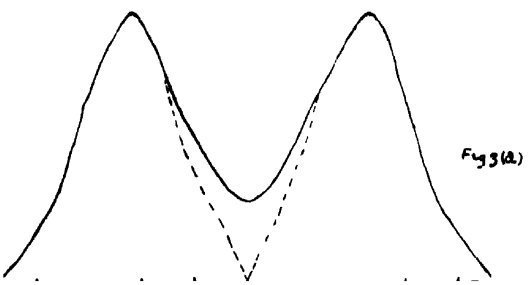


Fig. 3(b)
(*a*) Unextended, (*b*) Extended.

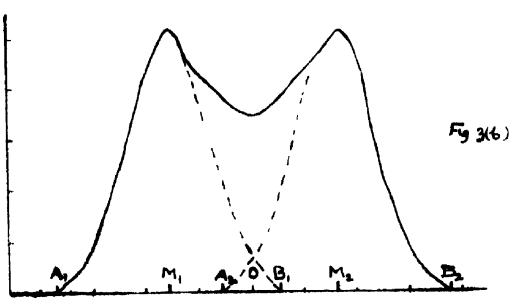


Fig. 3(a. b) Intensity distribution along (002) arc of X-ray photographs of parallel fibres :

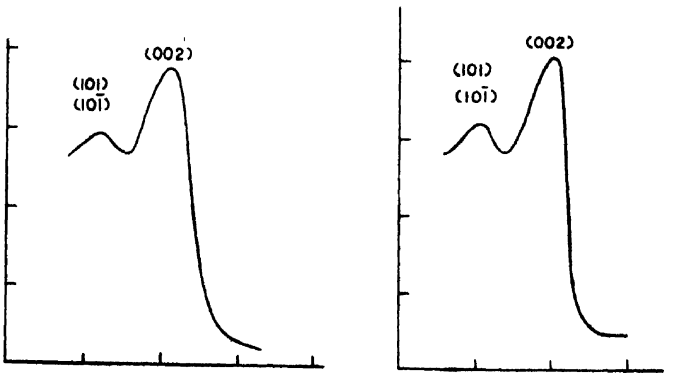


Fig. 4(a,b) Radial intensity distribution of X-ray photographs of roundomised fibres (*a*) Unextended (*b*) Extended

Taking the intensity $I(\alpha)$ of (002) arc for different values of angle(α) from the equator from Fig. 3, the Hermans [2] average angle of orientation has been calculated from the equation

$$\sin^2 \alpha_m = 2 \frac{\int_0^{\pi/2} I(\alpha) \sin^2\alpha \cos \alpha \, d\alpha}{\int_0^{\pi/2} I(\alpha) \cos \alpha \, d\alpha}$$

The integrals were evaluated graphically using a planimeter to measure the areas under the curves $I_{(\alpha)} \sin^2\alpha \cos \alpha$ Vs. α and $I_{(\alpha)} \cos\alpha$ Vs. α. The values are given in Table I

TABLE I

Sample	Spiral Angle (S)		Average Angle of orientation (α_m)		Cr. I	
	Unextended (U)	Extended (E)	Unextended (U)	Extended (E)	U	E
1	45°0′	30°0′	46°48′	37°0′	27.4%	30.3%
2	45°0′	32°30′	46°0′	38°0′	33.0%	36.3%

In Fig. 2, as a result of randomisation the contributions of the crystalline regions have been uniformly distributed over 360° giving rings of uniform intensity in place of the double humped arcs of Fig. 1. These photographs were radially scanned at intervals of 90° and a typical average intensity curve is shown in Fig. 4.

Considering the height *I*002 of the peak of (002) reflection above the background as the contribution of the crystalline regions and *I am*, the height of the minimum between the (002) and the composite (101 and 10I) peaks as the contribution of the amorphous portion, the following equation has been taken as the crystallinity index, CrI, after Segal *et al*, (1959) and Ingersoll (1946)

$$CrI = \frac{I002 - Iam}{I002} \times 100$$

The values of crystallinity index for the first series are given in Table I and those of second series in Table II. Each figure is based on observations on at least two pellets the difference between duplicates being on an average 1%.

Although, by condition of experiment, only part of the strain was retained in the fibre during X-ray examination, results in Table I clearly indicate that the extension of the fibre reduces the spiral angle (S), as observed by Stern (1957) the average angle (α_m) of orientation is also reduced but to a much smaller extent so as to increase the difference ($\alpha_m - S$) from 1° in the unextended state to 7° in the extended state. Since the spirality is reduced, this change in the ($\alpha_m - S$) may at first appear to be due to increased dispersion of crystallites wthin the microfibrils but Fig. 3 shows that the fibre elongation reduces the extent of the tail ends beyond the maximum points M_1 and M_2, that is, the va ue of $\frac{M_1A_1 + M_2B_2}{2}$ from 45° to 41° indicating according to Sisson (1935) reduced desperion of the crystallites within a microfibril or the microfibrils within a bundle of such. Actually, however, this depended on the method of calculating α_m, because in Fig. 3 when the dotted curves about the mean positions M_1 and M_2, representing half of the distribution of crystallites are considered it will be found that in obtaining a value of α_m for the left hand position $OM_1 A_1$ the tail end OB_1 beyond 0 (equator) was rejected while OA_2 on the left side of the equatior was taken into account this enhanced the value of α_m, as M_1 and M_2 come closer Moreover, the microfibrils near the edge of the spiral being not much inclined to the fibre axis also contribute towards increasing the value of αm

As regards crystallinity, Table I shows that the CrI of extended fibres was about 3% more than that of the unextended fibres for both samples. This may be explained if it is supposed that breaking extension not only reduces the angle which the microfibrils make with the fibre axis and the dispension amongst the existing crystallites within the microfibrils but also produces somewhat better alignment amongst the long chain molecules within the microfibrills and better planer regularity so as to assume a better lateral order, that is, to increase crystallinity. This is further verified when it is found from Table II that extension upto 10% do not produce any observable change in crystallinity index, the

Table II

Sample 3

Extension	Cr. I
0%	18%
10%	18%
20%	21%
Break	21.8%

the closeness of packing of the molecules being probably not sufficient to produce a permanent change in lateral order. On further extension however, CrI gradually increases, till at break the difference reaching the maximum value of 3.8% which is nearly the same as obtained for the previous series. Such increase in crystallinity on stretching has also been reported by M.Ruck-Florjancic (1960) *et al.* in case of regenerated fibres.

These results thus provided an explanation for the retention of about 4% even after drastic treatment as obtained by Stern and also for the properties introduced in the fibre by preconditioning under stress

ACKNOWLEDGMENT

The authors are thankful to Dr. A. C. Chakraborty, S.S.O., for many suggestions. Authors' thanks are is also due to Dr P. B. Sarkar, D.Sc., F.N.I., Director, for his keen interest in the work and to Indian Central Jute Committee for providing facilities for work and according permission to publish these results.

REFERENCES

Astbury, W. T., 1943. Textile Fibres under the X rays Imperial Chemical Industries Ltd., 27.

Hermans, P. H., 1946, Contrib to the Physics of Cellulose fibres, Amsterdam Elsevier Press p. 198.

Ingersoll, H. G., 1946, *J Appl Phys* **17**, 924.

Meredith, R., 1951, *J. Text Inst.*, **42** T275, T291

Preston, R. D., 1952, The molecular architecture of plant cell walls, 120. London, Chapman and Hall.

Ruck-orjancic, M., and Ruck, H., 1960, Svensk Papperstidn **63**, 845, *Rev. Textile Progress* **13**, 12

Stern, F., 1957, *J. Text. Inst.* **48**, T21

Stern, F., and Stout, H. P., 1954, **45**, T896.

Sisson, W. A., 1935, *Ind. Eng. Chem.*, **26**, 51

Segal, L., Creely, J. J., Manton, A. E., and Conrad, C. M., 1959, *Text. Res. J.*, **29**, 786.